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EXPERIMENTAL DETERMINATION OF THE $^1\Sigma^+$ STATE ELECTRIC DIPOLE MOMENT
FUNCTION OF CARBON MONOXIDE UP TO A LARGE INTERNUCLEAR SEPARATION

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SUMMARY

Experimental intensity information is combined with numerically obtained vibrational wave functions in a nonlinear least-squares fitting procedure to obtain the ground electronic state electric dipole moment function of carbon monoxide valid in the range of nuclear oscillation (0.87-1.91 Å) of about the $V = 38$ th vibrational level. Vibrational transition matrix elements are computed from this function for $\Delta V = 1, 2, 3$ with $V \leq 38$.

1. INTRODUCTION

Because of its large dissociation energy and the relatively high abundance of its constituent atoms, carbon monoxide in its several isotopic forms is observed in a variety of astrophysical sources. Because many of these sources are quite hot ($T > 7000$ K), the molecular emissions can be observed from highly excited vibrational levels that may or may not be in LTE. In this paper we report a determination of the electric dipole moment function (EDMF) of carbon monoxide that is accurate to a large internuclear separation. From this function we have calculated reliable estimates of the transition moments between highly excited vibrational levels. In addition to its application to astrophysical problems, this new information should be useful to experimenters who use the carbon monoxide laser in laboratory experiments.

Two basically different approaches can be used to determine EDMFs. One of them, direct ab initio calculation, offers the advantage that the range of internuclear separation is in principle not limited. However, this approach does not afford us estimates of errors and is most useful when experimental intensity information is not available or is limited. The second method, which involves the inversion of experimental intensity information, has the advantage that error estimates for the EDMF can be obtained from the errors in the experimental intensities. On the other hand, "experimental" determinations of EDMFs (with the notable exception of the HF and HCl molecules (ref. 1)) have been limited to the lowest few vibrational levels and, therefore, to smaller ranges of the vibrational coordinate.

In this paper we report the determination of the EDMF of carbon monoxide via the inversion of experimental intensity information. In a previous paper (ref. 2) we reported a hybrid EDMF which reproduces the experimental intensity measurements

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(0-1, 0-2, 0-3, 0-4 vibrational bands) near equilibrium and ab initio calculations of the EDMF at a very large internuclear separation. Therefore, up to this report the EDMF of carbon monoxide has been well determined experimentally for a range of nuclear oscillation somewhat inside the classical turning points of the fourth vibrational level (1.01-1.30 Å). For the analysis reported in this paper we have incorporated an extensive set of new experimental information on the intensity ratios of the $\Delta V = 1, 2, 3$ ($V \leq 38$) vibrational transitions. This work, therefore, extends by more than a factor of 3 the range (0.87-1.91 Å) over which the EDMF of carbon monoxide has been experimentally determined.

In the following section we briefly discuss the experiments that were done to obtain the experimental intensity ratios of vibrational transitions which involve highly excited vibrational levels. Next, these ratios taken with the absolute vibrational intensities of the 0-V ($V = 1-4$) bands are "inverted" to yield the ground electronic state EDMF for the range of internuclear separations cited above. Finally, vibrational matrix elements of the derived EDMF are reported.

2. EXPERIMENTAL INTENSITY RATIOS

The carbon monoxide emission spectra we used to obtain intensity ratios were produced using a discharge source constructed in Bonn and recorded with the high-information Fourier transform interferometer (ref. 3) at the Laboratoire d'Infrarouge. We give here only a brief discussion of the spectral data and the procedure for obtaining intensity ratios from these data since these points are discussed in detail in another report (ref. 4).

The radiation source was basically a gas laser with one of the cavity mirrors removed and one retained so as to increase the power delivered along the axis of the source to the interferometer. The entire optical path of the source-interferometer system, other than the source itself, was evacuated. The two emission spectra used for this work include the following sets of vibrational transitions: $\Delta V = 1, 2$ (V up to 37) and $\Delta V = 3$ ($V = 12-38$). Rovibrational emission intensities were measured for three isotopic species in the isotopically normal sample of carbon monoxide; however, for the purpose of this work only the information for $^{12}\text{C}^{16}\text{O}$ was used for the EDMF determination. Since these spectra are well described with an extended set of Dunham constants (ref. 5) it was possible to select lines for the intensity work that were not sensibly overlapped.

For the purpose of further discussion we give the equation for the purely vibrational transition moment for emission ($-\Delta V$):

$$T(V, -\Delta V) = \frac{V!}{(V - \Delta V)! \Delta V!} T(0, \Delta V) H(V, -\Delta V) \quad (1)$$

where we represent the squared matrix element of the EDMF (the transition moment) by $T(V, -\Delta V) = |\langle V | M | V - \Delta V \rangle|^2$, M represents the EDMF, V is the upper vibrational state quantum number, and $H(V, -\Delta V)$ the anharmonicity factor. It was possible to determine for each vibrational transition, ($V, -\Delta V$), the quantity $N(V)H(V, -\Delta V)$, where $N(V)$ is the population of the V th vibrational level (ref. 4). This information was obtained from the intercept on a semilog plot of the rovibrational line intensities versus the rotational energy. The procedure we actually used (ref. 4) to obtain line intensities from the line-emission measurements follows generally a method used previously (refs. 6 and 7) but modified somewhat to accommodate the particular source we used.

The standard deviations obtained from the least-squares determinations of individual $N(V)H(V,-\Delta V)$ values are of the order of 3% to 5% for most of the vibrational levels, but can be as high as 10% for (a) the highest levels (because the associated transitions have very low intensities) and (b) the lowest levels ($V \leq 3$), for which the non-uniformity of temperature conditions along the axis of the source (ref. 4) is relatively more difficult to correct.

For the purpose of the nonlinear determination of the EDMF we form ratios of $N(V)H(V,-\Delta V)$ with the same V in the numerator and denominator so that the unknown populations $N(V)$ will cancel:

$$\frac{T(V,\Delta V)}{T(V,-1)} = \frac{(V-1)!T(0,\Delta V)H(V,-\Delta V)}{\Delta V!(V-\Delta V)!T(0,1)H(V,-1)} \quad (2)$$

We computed the right-hand side of equation (2) for $\Delta V = 2$ and $\Delta V = 3$ using the experimentally determined values of $N(V)H(V,-\Delta V)$ and the appropriate $T(0,\Delta V)$ values taken from previously determined absorption experiments; these quantities are tabulated in table 1.

3. VIBRATIONAL WAVE FUNCTIONS

The two basic ingredients for our nonlinear least-squares determination of the EDMF are the experimental intensity ratios and the appropriate vibrational wave functions. We computed the wave functions by numerically solving the radial Schrodinger equation (ref. 8) using an RKR vibrational potential (ref. 9) based on the Dunham coefficients reported by Dale et al. (ref. 10). The latter coefficients predict vibrational energies for the highest levels ($V = 30-38$) that are different by about $10-20 \text{ cm}^{-1}$ from the predictions of more recent work (ref. 5), however, this discrepancy should have little effect on the computed vibrational wave functions. The wave functions were computed for a range of bond distances between 0.45 \AA and 3.00 \AA at equally spaced intervals (0.002 \AA). The interval size was chosen so as to resolve the rapid oscillations in the wave functions of the highest levels, and the range for the bond distance extends well beyond the classical turning points of the $V = 38$ th vibrational level. The probability for nuclear position shown in figure 1 for the $V = 0$ and $V = 34$ state indicates the large range of nuclear oscillations involved with the vibrational levels considered in this report. As can be seen in figure 1, the probability for the lowest level ($V = 0$) is peaked near the equilibrium internuclear separation ($R_e = 1.128 \text{ \AA}$), on the other hand, the probability for one of the highest levels ($V = 34$) is peaked near the classical turning points.

For the purpose of the least-squares fitting we computed 1,276 points each for 39 vibrational wave functions and stored this information as a permanent file on a removable hard-disk pack used in conjunction with a CDC 7600 computer. Next, products of upper and lower state wave functions (1,276 points each) were computed for all of the transitions considered for a particular fit, these products (weighting functions) were stored for repeated use in the iterative least-squares determination of the EDMF. Vibrational weighting functions are shown for $V = 34 \rightarrow V = 35$ (fig. 2); $V = 34 \rightarrow V = 36$ (fig. 3), and $V = 34 \rightarrow V = 37$ (fig. 4). The patterns are fairly complex but indicate the regions of R where the slowly varying EDMF is most (least) heavily weighted by a particular transition. For example, the region near $R = 1.65 \text{ \AA}$ of the $V = 34 \rightarrow V = 35$ transition (fig. 2) has, on average, zero weight, whereas the region of the classical turning points is heavily weighted. Given ΔV sequences have patterns similar to those shown in figures 2-4, with regions of zero weighting

changing smoothly with V . We have not made systematic checks but we believe the vibrational weighting functions represented by our data set "probe" the EDMF more or less equitably over the range of R considered.

4. THE ELECTRIC DIPOLE MOMENT FUNCTION

In this section we incorporate measured vibrational intensities and computed vibrational wave functions into a nonlinear least-squares determination of the parameters in a Padé approximate (ref. 11) representation of the EDMF. Figure 5 is a flow diagram for our overall computational procedure. The adjustment of parameters $A(1)$ in the Padé representation was controlled with a code written by Gill et al. (ref. 12), and the matrix elements of the EDMF were computed via Simpson's rule. The fit incorporated previously measured absorption transition moments and the moment ratios for the highly excited vibrational levels. We initially made fits assuming the $0 \rightarrow V$ ($V = 1, 2, 3, 4$) transition matrix elements suggested in reference 2, but found a better overall fit (by about 2% in the ratios) if we used a $V = 0 \rightarrow V = 1$ transition moment 2.2% smaller than that suggested by Chackerian and Tipping (ref. 2); this latter value is in agreement with the measurements (not the calculations) of Chackerian et al. (ref. 13). The form of the Padé approximate we used to represent the EDMF is expressed in terms of the dimensionless coordinate $\xi = (R - R_e)/(R_e)$ with $R_e = 1.1283226 \text{ \AA}$

$$M(\xi) = \frac{-0.122706[1 + A(1)\xi + A(2)\xi^2 + A(3)\xi^3]}{1 + A(4)\xi^1 + A(5)\xi^2 + A(6)\xi^3 + A(7)\xi^6} \quad (3)$$

where the equilibrium moment is held fixed and equal to the leading term in the numerator. It was found that this representation for $M(\xi)$ gives a better fit to the data than one that decreases with asymptotically large ξ as ξ^{-4} . This is not surprising since the data are not sensitive to this region, the EDMF reported here should, therefore, not be used outside the range of internuclear separation of 0.87-1.91 \AA . The parameters $A(i)$ that gave the best fit to the data are given in table 2, and the transition moments computed using these parameters are compared with the experimental values in table 1. As can be seen, the fit is good: the 57 emission transition moment ratios are fitted to about $\pm 8\%$ and the absorption transition moments are fitted to about $\pm 2\%$.

In figure 6 we show a comparison between the EDMF determined in this work and the previous numerical computation of Chackerian and Tipping (ref. 2). The two functions differ by about 2% and 14%, respectively, at the outer turning points of the $V = 21$ st and $V = 38$ th levels. These rather small differences in the two EDMFs translate into much larger differences in the computed transition moment ratios. This can be seen in figures 7 and 8, where the computations of the $\Delta V = 2$ and $\Delta V = 3$ ratios are compared using the two different functions. The discrepancy for $\Delta V = 2$ increases to 59% at $V = 37$, for $\Delta V = 3$, the discrepancy ranges from -55% at $V = 12$ to about 200% at $V = 38$.

Finally, in table 3, we give the vibrational transition moments that best represent the data. The transition moments were computed using the numerically obtained wavefunctions and the EDMF we have determined. We hope to compute soon reliable rotational wavefunctions for these highly excited vibrational levels so that the rotational Herman-Wallis factor may be obtained.

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TABLE 1.- COMPARISON OF OBSERVED AND CALCULATED
TRANSITION MOMENTS

V	V-ΔV	Transition moment ratios		
		Experimental	Calculated	Calc/Exper
3	1	0.4276(-2)	0.4055(-2)	0.95
4	2	.6415(-2)	.6237(-2)	.97
5	3	.8470(-2)	.8511(-2)	1.01
6	4	.1059(-1)	.1087(-1)	1.03
7	5	.1283(-1)	.1330(-1)	1.04
8	6	.1591(-1)	.1581(-1)	.99
9	7	.1818(-1)	.1838(-1)	1.01
10	8	.2085(-1)	.2102(-1)	1.01
11	9	.2411(-1)	.2371(-1)	.98
12	10	.2652(-1)	.2647(-1)	1.00
13	11	.2952(-1)	.2928(-1)	.99
14	12	.3265(-1)	.3216(-1)	.99
19	17	.4609(-1)	.4762(-1)	1.03
20	18	.4865(-1)	.5099(-1)	1.05
21	19	.5227(-1)	.5448(-1)	1.04
22	20	.5711(-1)	.5812(-1)	1.02
23	21	.6042(-1)	.6191(-1)	1.03
24	22	.6643(-1)	.6588(-1)	.99
25	23	.7141(-1)	.7006(-1)	.98
26	24	.7438(-1)	.7448(-1)	1.00
27	25	.8135(-1)	.7916(-1)	.97
28	26	.8361(-1)	.8416(-1)	1.01
29	27	.8845(-1)	.8951(-1)	1.01
30	28	.9634(-1)	.9526(-1)	.99
31	29	.1017	.1014	1.00
32	30	.1104	.1082	.98
33	31	.1151	.1155	1.00
34	32	.1236	.1235	1.00
35	33	.1312	.1323	1.00
36	34	.1391	.1420	1.02
37	35	.1490	.1528	1.03
12	9	.5743(-3)	.6144(-3)	1.07
13	10	.7540(-3)	.7785(-3)	1.03
14	11	.9464(-3)	.9676(-3)	1.02
15	12	.1196(-2)	.1183(-2)	.99
16	13	.1436(-2)	.1425(-2)	.99
17	14	.1743(-2)	.1695(-2)	.97
18	15	.2077(-2)	.1994(-2)	.96
19	16	.2480(-2)	.2323(-2)	.94
20	17	.2828(-2)	.2682(-2)	.95
21	18	.3134(-2)	.3073(-2)	.97
22	19	.3580(-2)	.3496(-2)	.98
23	20	.4057(-2)	.3953(-2)	.97
24	21	.4488(-2)	.4445(-2)	.99
26	23	.5594(-2)	.5544(-2)	.99
27	24	.6122(-2)	.6157(-2)	1.01
28	25	.6612(-2)	.6817(-2)	1.03

TABLE 1.- CONCLUDED

V	V-ΔV	Transition moment ratios		
		Experimental	Calculated	Calc/Exper
29	26	0.7191(-2)	0.7530(-2)	1.05
30	27	.7880(-2)	.8303(-2)	1.05
31	28	.8700(-2)	.9143(-2)	1.05
32	29	.9679(-2)	.1006(-1)	1.04
33	30	.1096(-1)	.1107(-1)	1.01
34	31	.1239(-1)	.1218(-1)	.98
35	32	.1398(-1)	.1342(-1)	.96
36	33	.1598(-1)	.1480(-1)	.93
37	34	.1770(-1)	.1638(-1)	.93
38	35	.1853(-1)	.1820(-1)	.98
Transition moments, Debye ²				
1	0	0.1150(-1)	0.1131(-1)	0.98
2	0	.4356(-4)	.4449(-4)	1.02
3	0	.1840(-6)	.1829(-6)	.99
4	0	.4064(-9)	.4066(-9)	1.00

TABLE 2.- EDMF^a COEFFICIENTS
DETERMINED FOR EQUATION (3)

i	A(i)
1	-28.82477
2	4.22116
3	-222.8626
4	0.265928
5	8.85457
6	1.14562
7	9.71303

^aValid between about 0.866
and 1.927 Å.

TABLE 3. VIBRATIONAL TRANSITION MOMENTS,
T(V,- Δ V), DEBYE²

V/ Δ V	1	2	3
1	0.1131(-1)		
2	.2254(-1)	0.4449(-4)	
3	.3370(-1)	.1367(-3)	0.1829(-6)
4	.4478(-1)	.2793(-3)	.8089(-6)
5	.5577(-1)	.4747(-3)	.2221(-5)
6	.6667(-1)	.7247(-3)	.4849(-5)
7	.7749(-1)	.1031(-2)	.9206(-5)
8	.8821(-1)	.1395(-2)	.1588(-4)
9	.9884(-1)	.1817(-2)	.2555(-4)
10	.1094	.2299(-2)	.3892(-4)
11	.1198	.2842(-2)	.5679(-4)
12	.1302	.3445(-2)	.7997(-4)
13	.1404	.4111(-2)	.1093(-3)
14	.1505	.4840(-2)	.1456(-3)
15	.1605	.5634(-2)	.1899(-3)
16	.1704	.6492(-2)	.2428(-3)
17	.1801	.7418(-2)	.3053(-3)
18	.1897	.8413(-2)	.3783(-3)
19	.1991	.9480(-2)	.4624(-3)
20	.2083	.1062(-1)	.5586(-3)
21	.2172	.1184(-1)	.6676(-3)
22	.2260	.1313(-1)	.7901(-3)
23	.2345	.1451(-1)	.9268(-3)
24	.2426	.1598(-1)	.1079(-2)
25	.2504	.1755(-1)	.1246(-2)
26	.2579	.1921(-1)	.1430(-2)
27	.2649	.2097(-1)	.1631(-2)
28	.2715	.2285(-1)	.1851(-2)
29	.2776	.2484(-1)	.2090(-2)
30	.2830	.2696(-1)	.2350(-2)
31	.2879	.2921(-1)	.2632(-2)
32	.2921	.3161(-1)	.2939(-2)
33	.2955	.3414(-1)	.3271(-2)
34	.2982	.3684(-1)	.3632(-2)
35	.2999	.3969(-1)	.4025(-2)
36	.3008	.4271(-1)	.4453(-2)
37		.4591(-1)	.4922(-2)
38			.5442(-2)

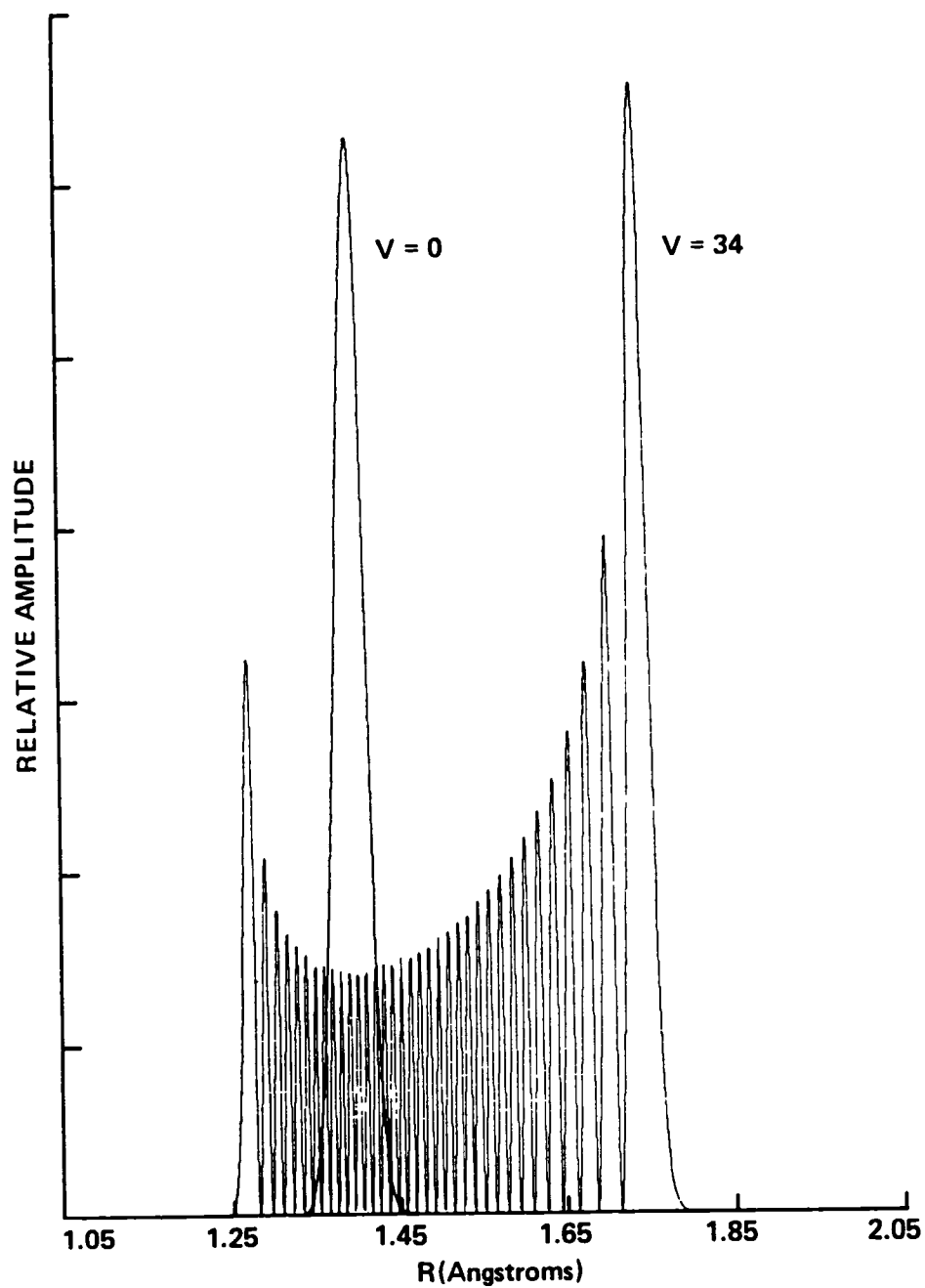


Figure 1.- Probability amplitude for nuclear position in the $V = 0$ and $V = 34$ vibrational states. For proper relative scaling multiply the vertical component of the $V = 0$ graph by 2.5.

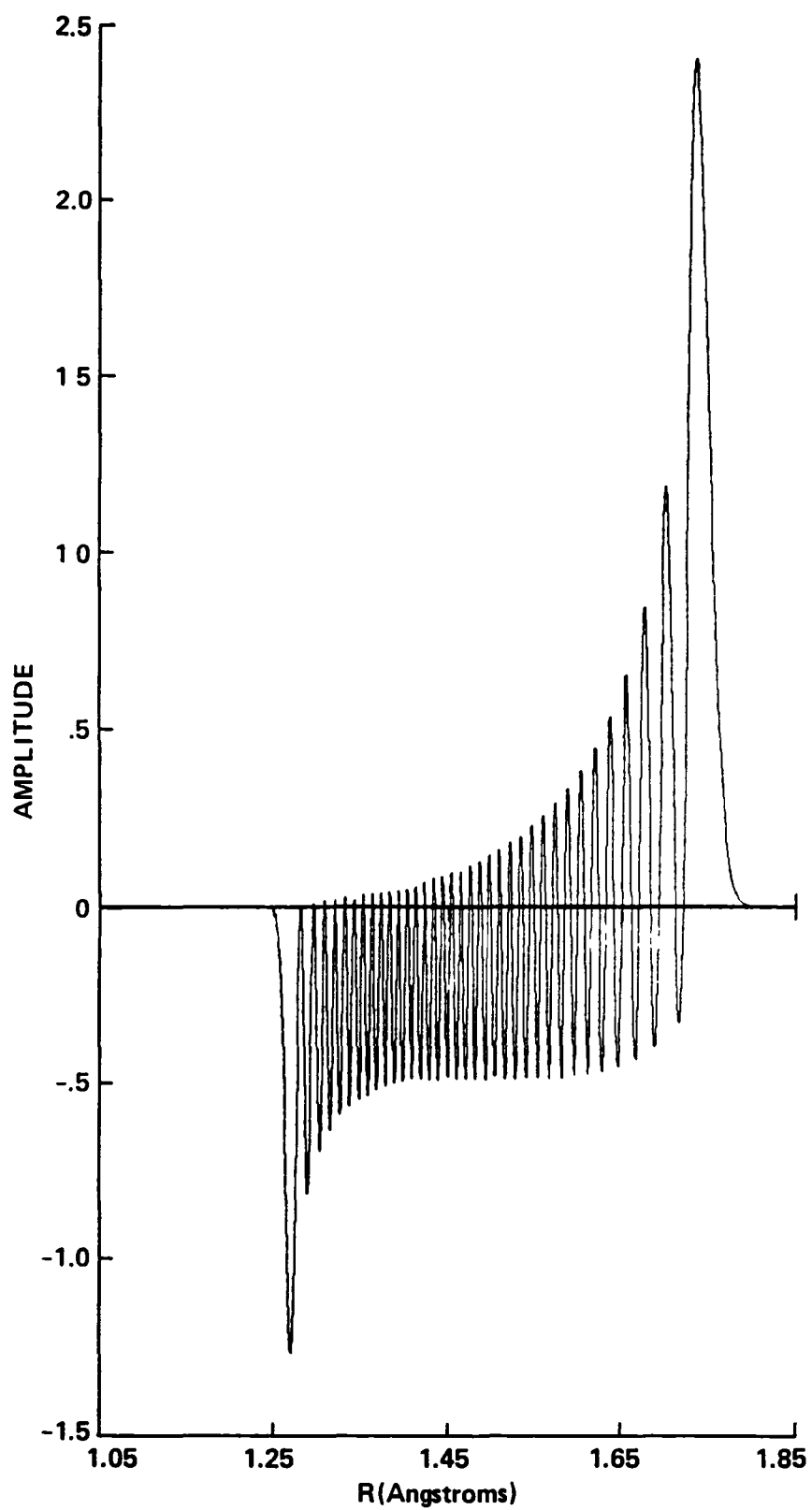


Figure 2.- Vibrational weighting function for the $V = 34 \rightarrow V = 35$ transition.

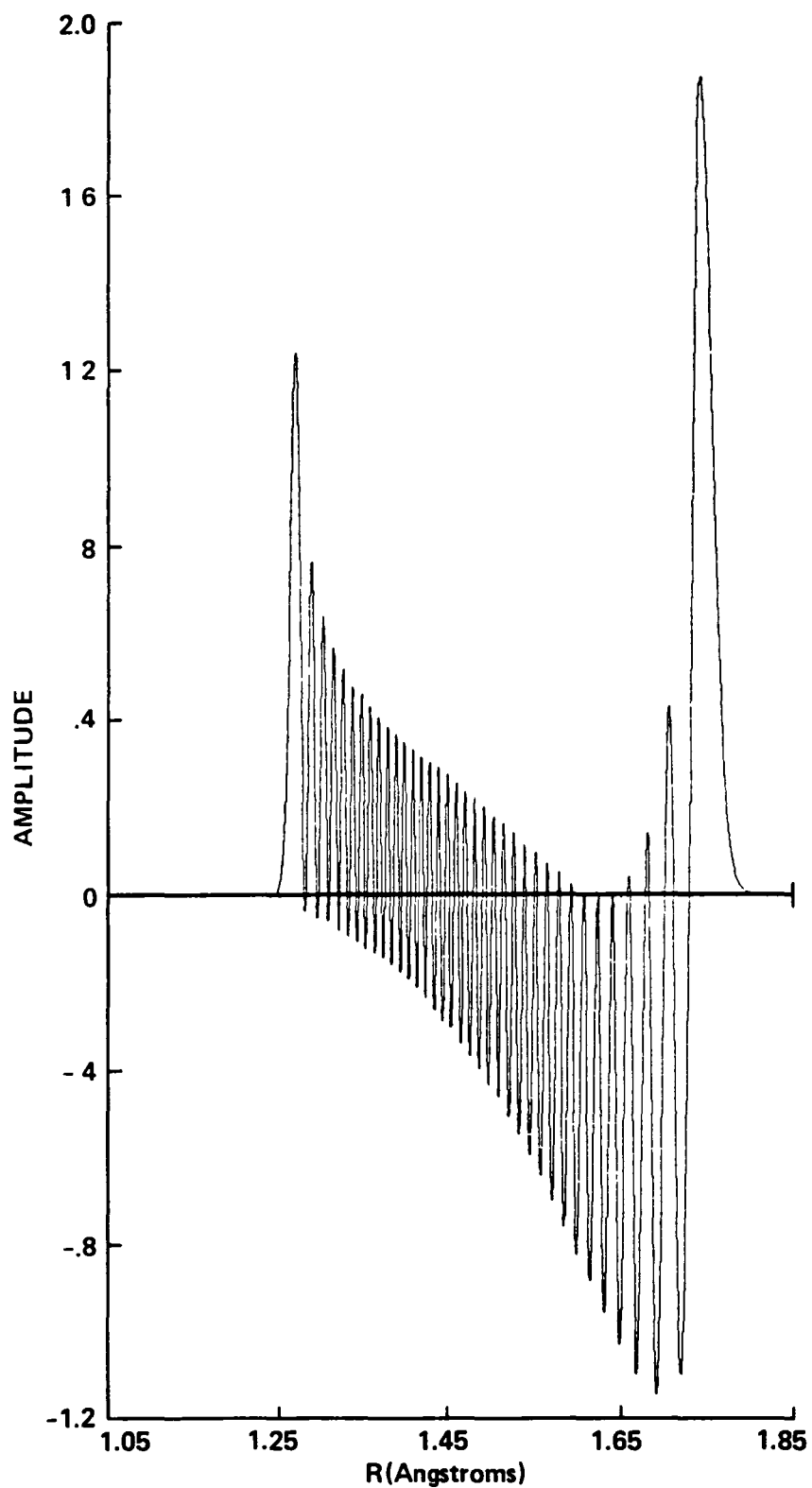


Figure 3.- Vibrational weighting function for the $V = 34 \rightarrow V = 36$ transition.

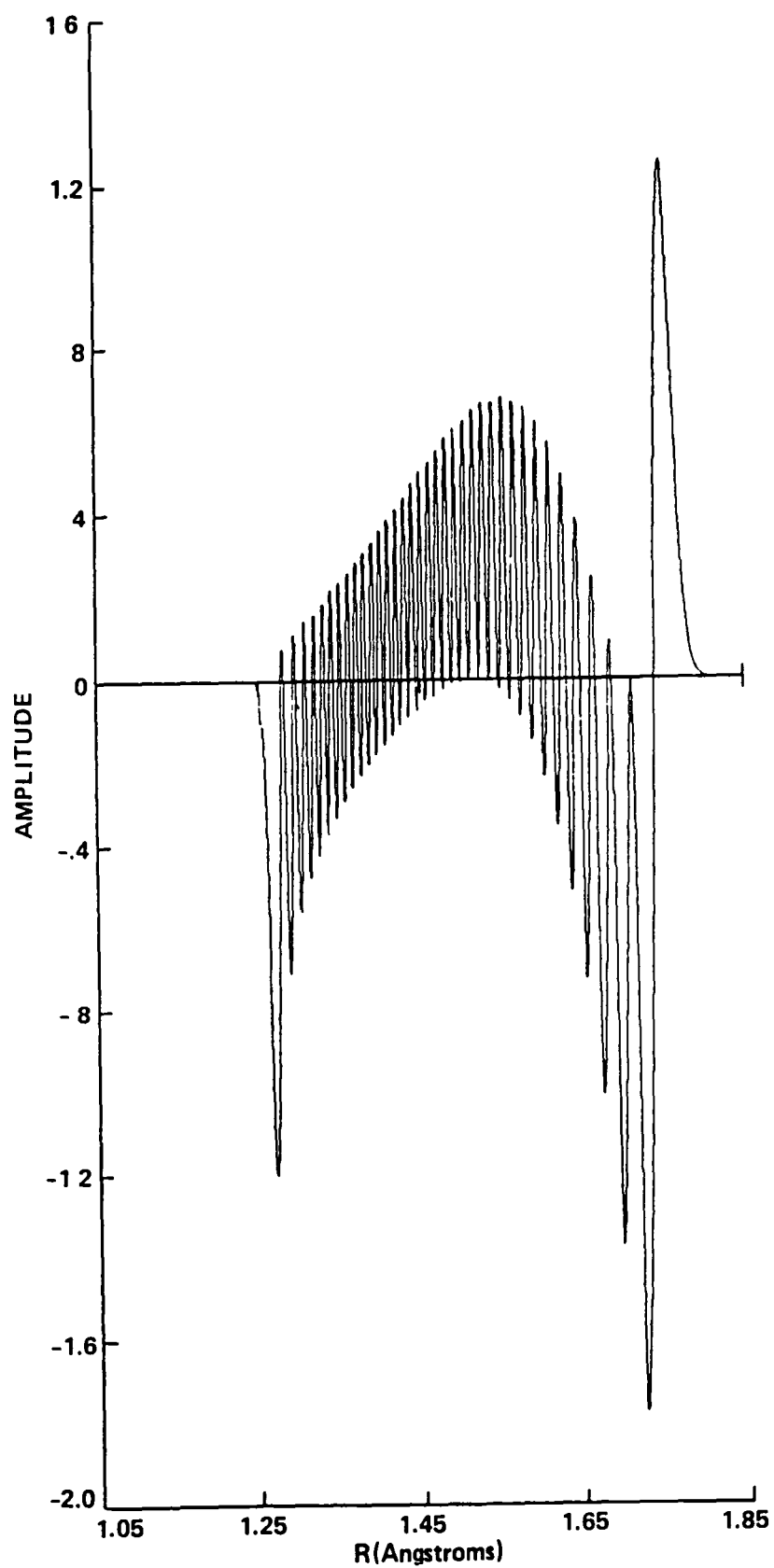


Figure 4.- Vibrational weighting function for the $V = 34 \rightarrow V = 37$ transition.

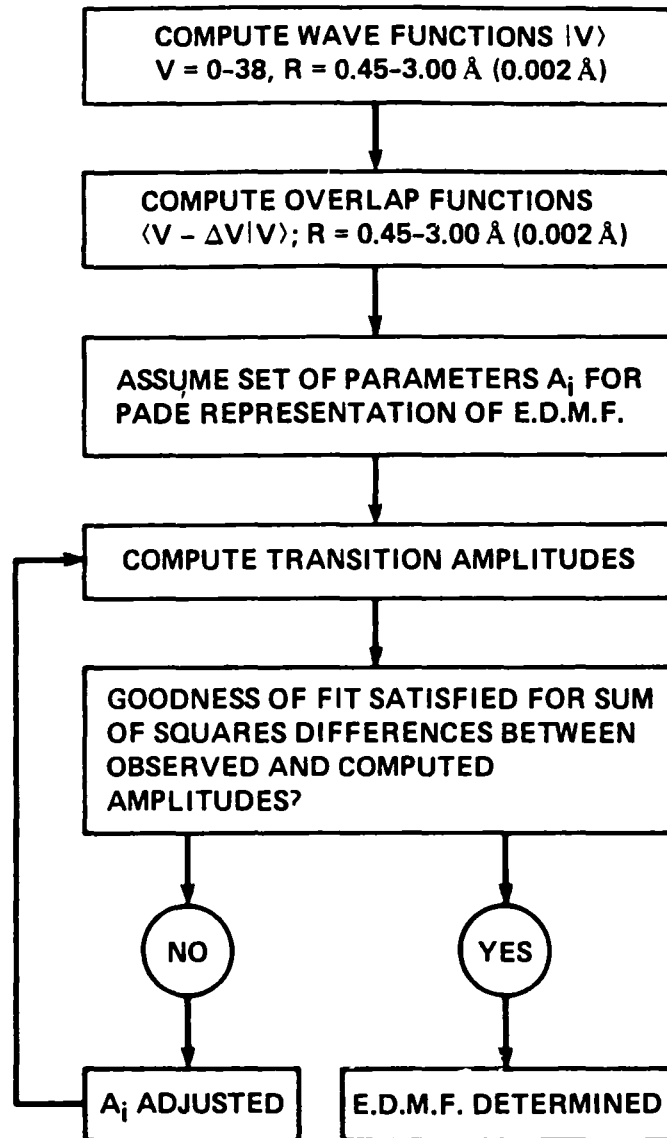


Figure 5.- Computational flow diagram for EDMF determination.

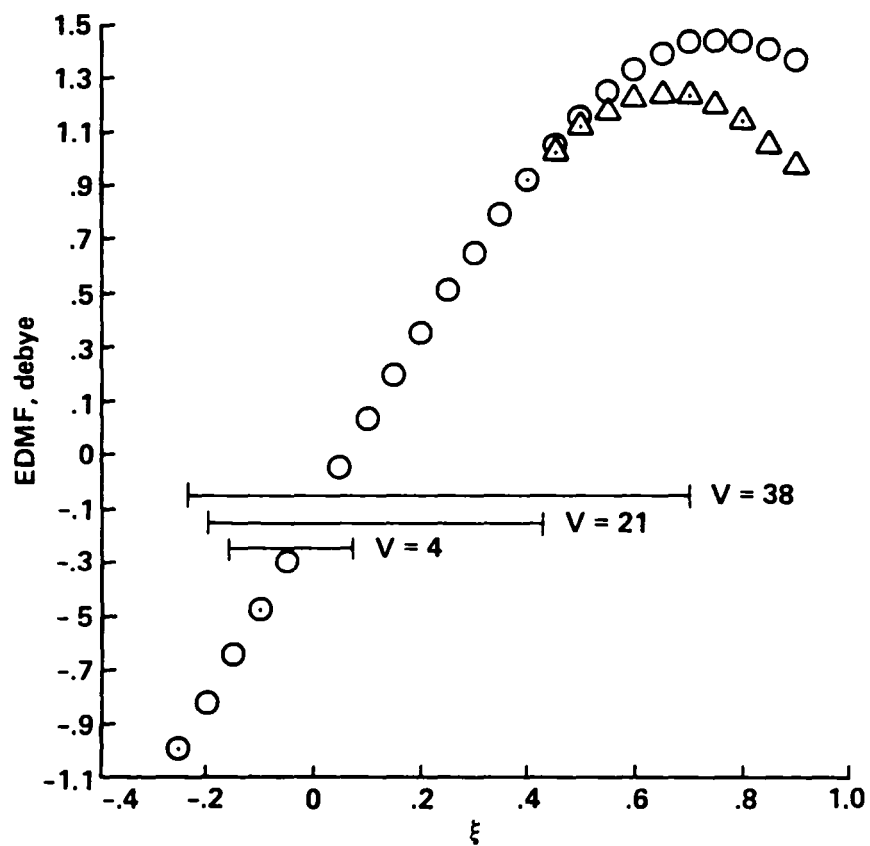


Figure 6.- EDMF versus dimensionless coordinate ξ . \odot , this work; \square , reference 2. Horizontal bars indicate the range of classical oscillation.

Fig. 6

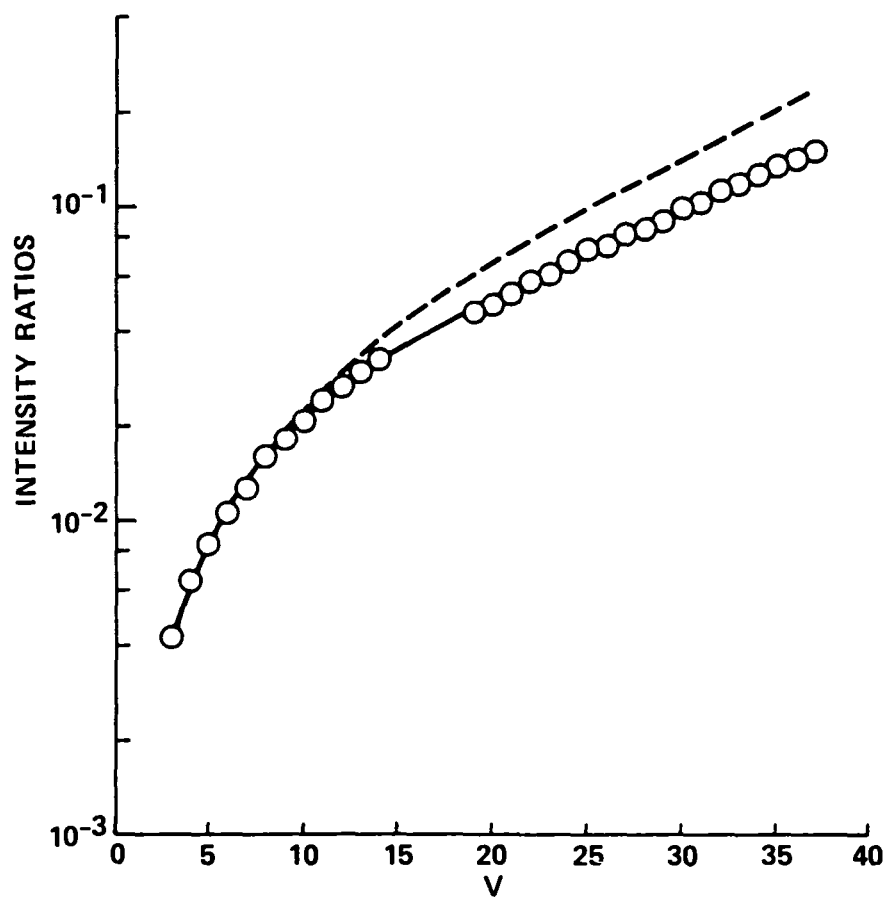


Figure 7.- Transition moment ratios (see equation (2)) versus V for $\Delta V = 2$.
 \odot , experimental; —, this work; ---, reference (2).

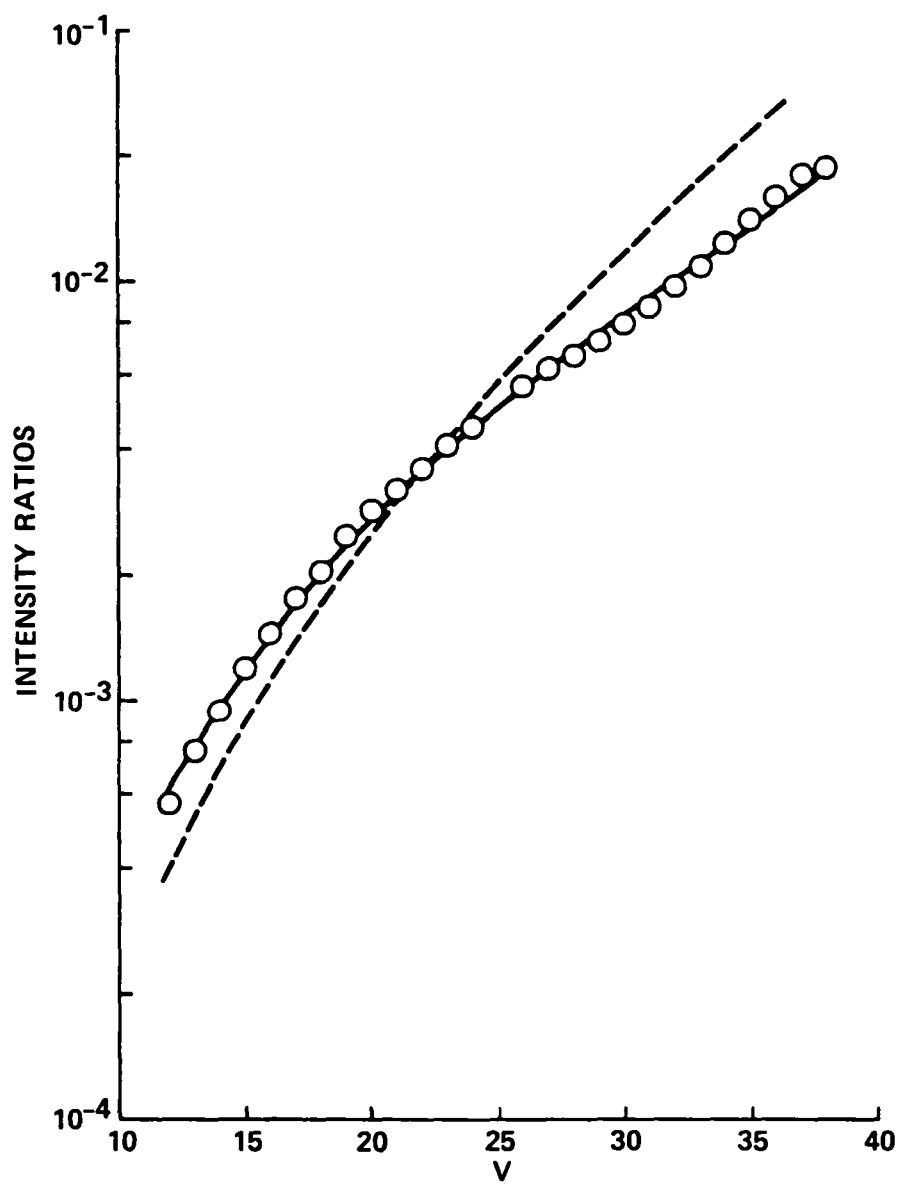


Figure 8.- Transition moment ratios (see equation (2)) versus V for $\Delta V = 3$.
 \odot , experimental; —, this work; ---, reference (2).

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